

## ***Supporting Information***

### **Phosphorous Doped Two-dimensional CoFe<sub>2</sub>O<sub>4</sub> Nanobelt Decorated with Ru Nanoclusters and Co-Fe Hydroxide as Efficient Electrocatalysts Toward Hydrogen Generation**

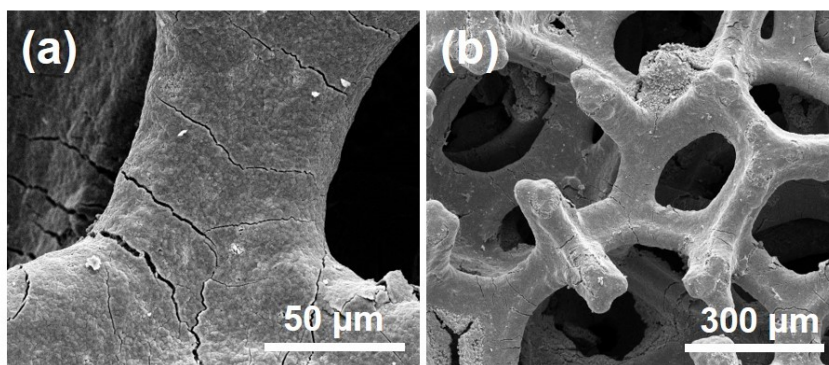
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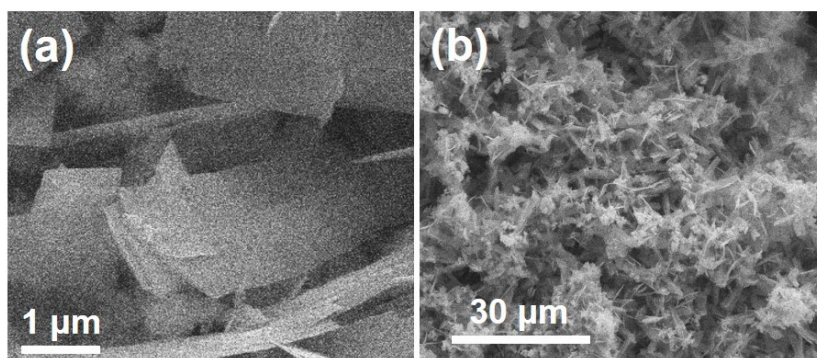
2. Key Laboratory of Eco-chemical Engineering, Taishan Scholar Advantage and Characteristic Discipline Team of Eco Chemical Process and Technology, Ministry of Education, Laboratory of Inorganic Synthesis and Applied Chemistry, College of Chemistry and Molecular Engineering, Qingdao University of Science and Technology, Qingdao 266042, P. R. China E-mail: splswzx@qust.edu.cn,

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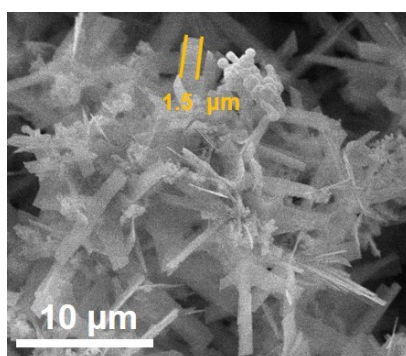
4. Key Laboratory of Advanced Energy Materials Chemistry (Ministry of Education), College of Chemistry, Nankai University, Tianjin 300071, China



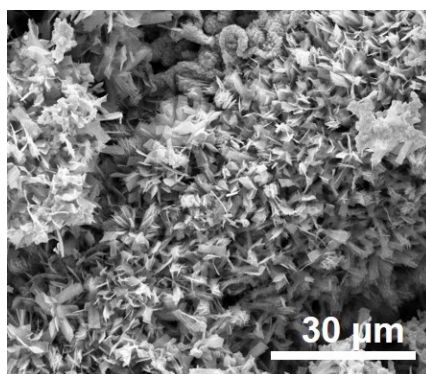
**Figure S1** SEM images of iron foam with different magnification.



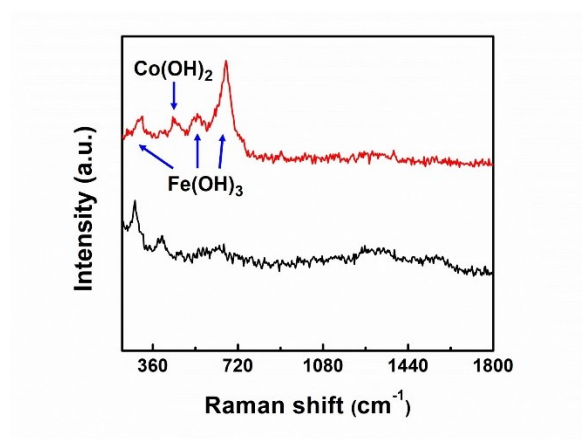
**Figure S2** SEM images of the product after the hydrothermal reaction with different magnification.



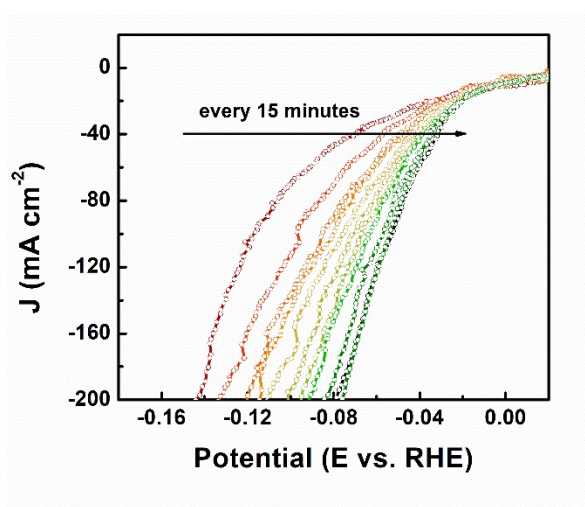
**Figure S3** SEM image of the product after the hydrothermal reaction.



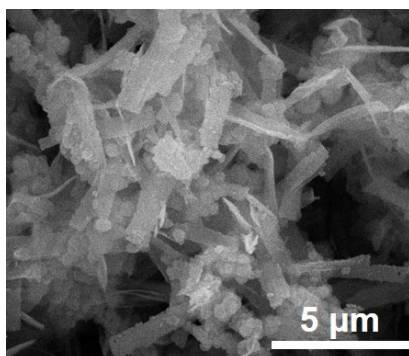
**Figure S4** SEM image of Ru/P-CoFe<sub>2</sub>O<sub>4</sub>/IF.



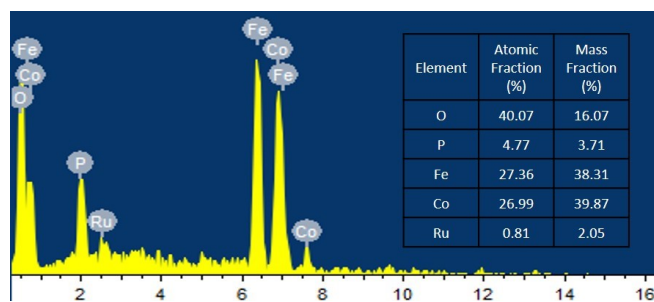
**Figure S5** Raman spectra of CoFeO<sub>x</sub>H<sub>y</sub>-Ru/P-CoFe<sub>2</sub>O<sub>4</sub>/IF (red line) and Ru/P-CoFe<sub>2</sub>O<sub>4</sub>/IF (black line).



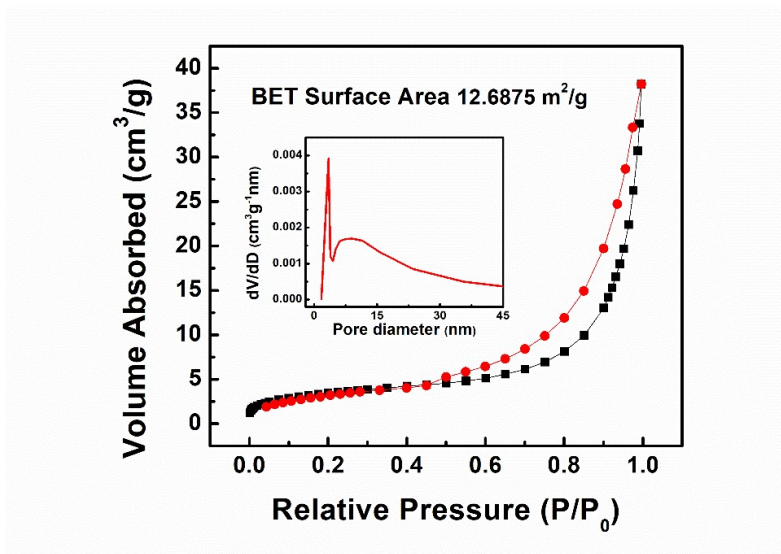
**Figure S6** HER polarization curves of as prepared catalyst tested every 15 minutes.



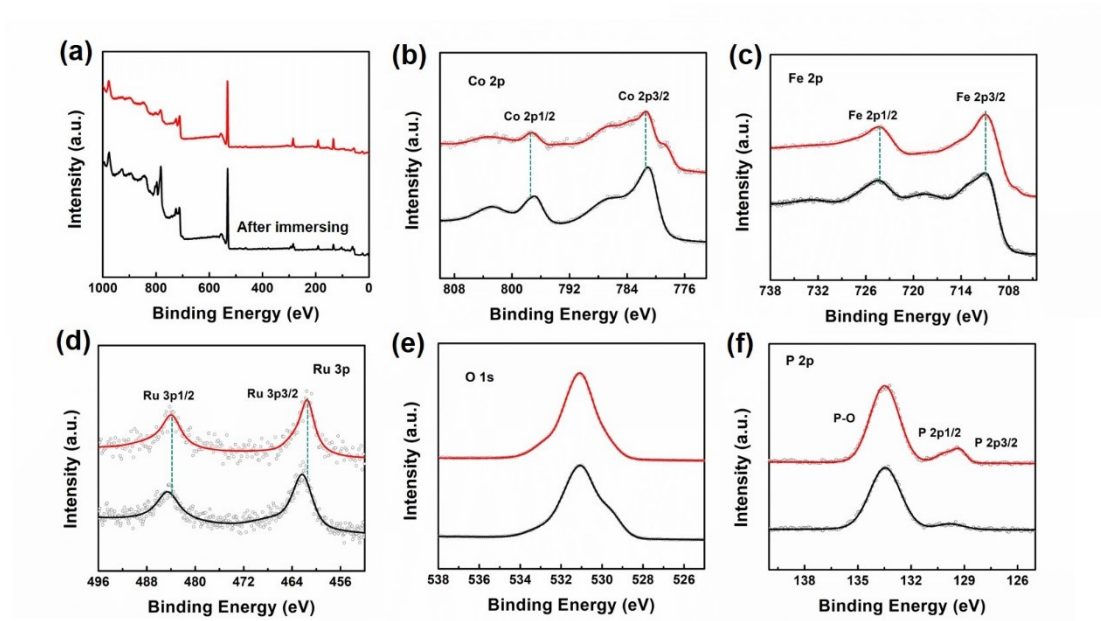
**Figure S7** SEM image of pre-catalyst after soaking in water.



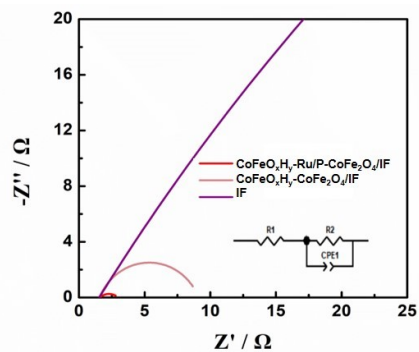
**Figure S8** the EDS mappings  $\text{CoFeO}_x\text{H}_y\text{-Ru/P-CoFe}_2\text{O}_4/\text{IF}$ .



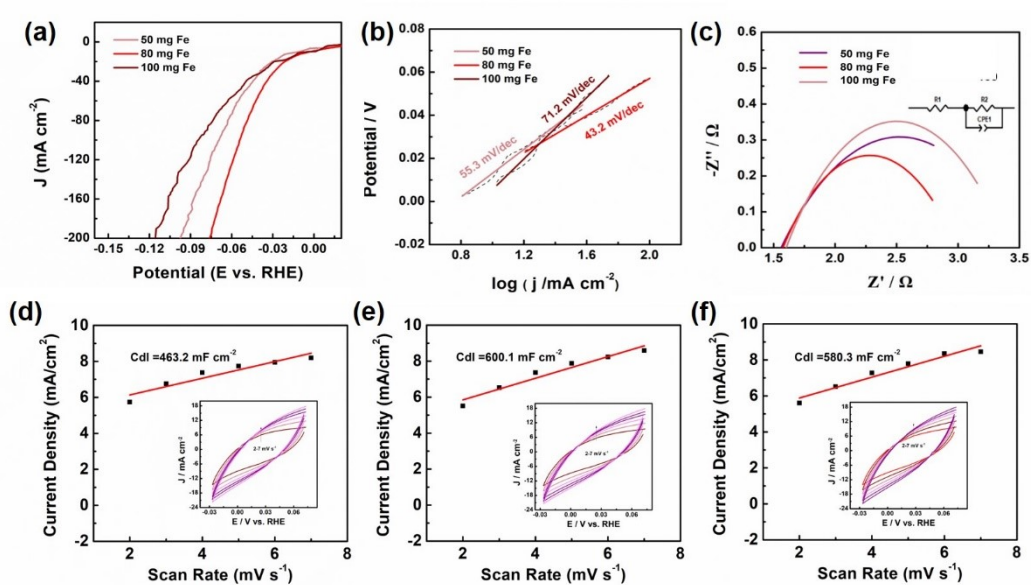
**Figure S9**  $N_2$  sorption isotherms and pore size distribution curve of  $CoFeO_xH_y$ -Ru/P- $CoFe_2O_4$ /IF.



**Figure S10** XPS survey spectrum (a) high-resolution of Co 2p (b), Fe 2p (c), Ru 3p (d), O 1s (e) and P 2p (f) of the designed Ru/P- $CoFe_2O_4$ /IF (red line) and  $CoFeO_xH_y$ -Ru/P- $CoFe_2O_4$ /IF (black line).

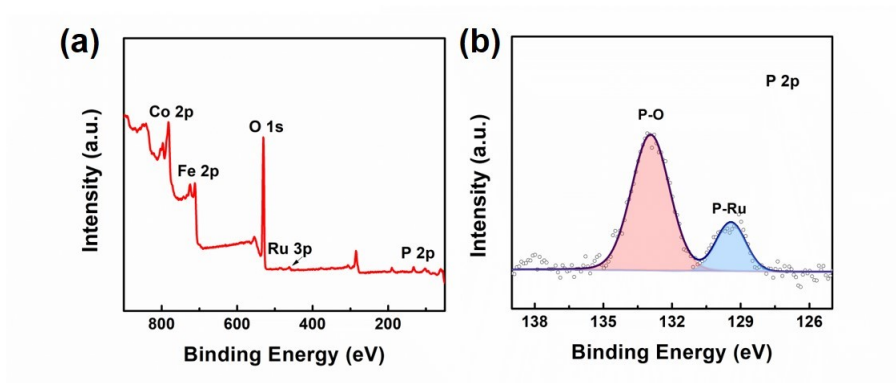


**Figure S11** Nyquist plots of obtained catalysts in 1M KOH.

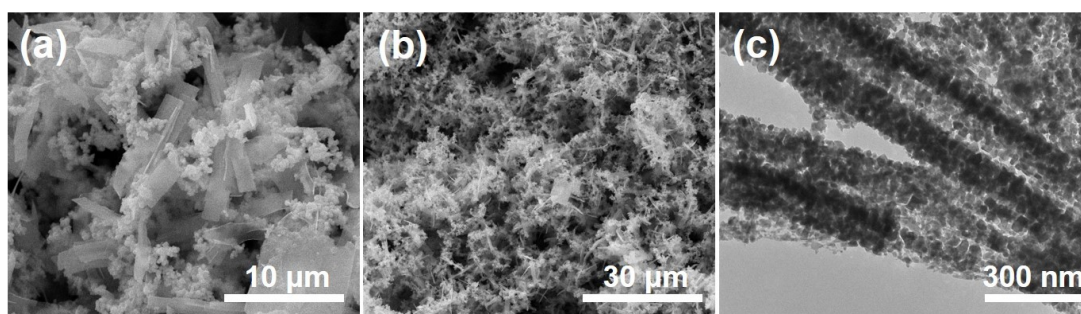


**Figure S12** Electrochemical measurements: (a) HER polarization curves, (b) Tafel slopes, (c) Nyquist plots, (d-f) linear fitting of scan rates with capacitive current densities (inset is CV curves under different scan rate) of obtained catalysts in 1M KOH.

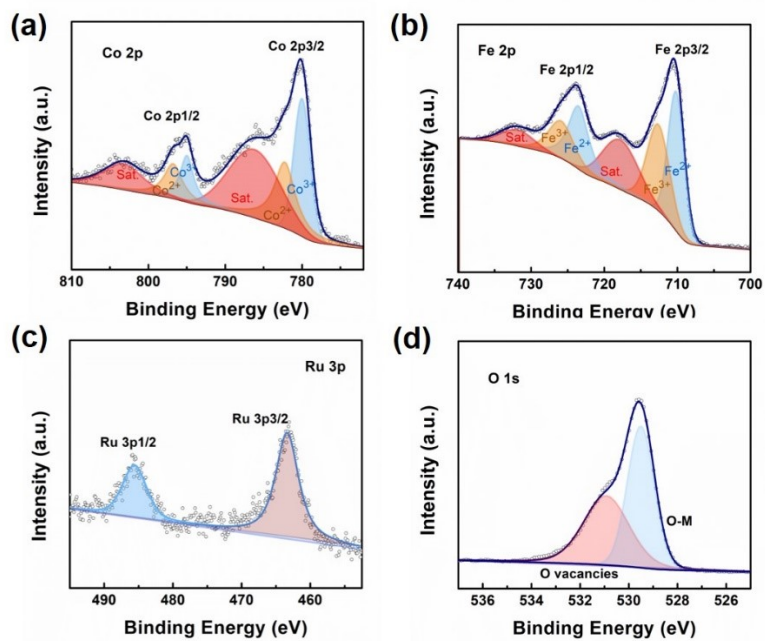




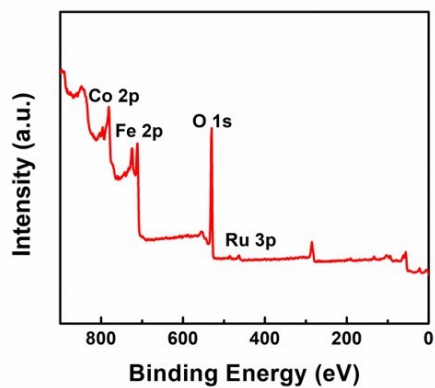
**Figure S13** XPS survey spectrum (a) and high-resolution of P 2p (b) of the designed  $\text{CoFeO}_x\text{H}_y\text{-Ru/P-CoFe}_2\text{O}_4/\text{IF}$  after long-time stability test.



**Figure S14** (a, b) SEM image of  $\text{RuO}_2/\text{CoFe}_2\text{O}_4$  with different magnification. (c) TEM image of  $\text{RuO}_2/\text{CoFe}_2\text{O}_4$ .

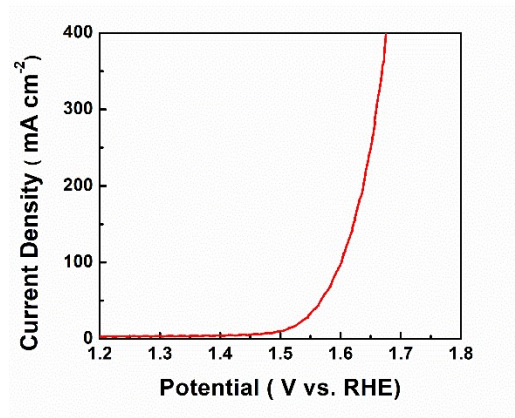


**Figure S15** XPS survey spectrum high-resolution of Co 2p (a), Fe 2p (b), Ru 3p (c) and O 1s (d) of the designed RuO<sub>2</sub>/CoFe<sub>2</sub>O<sub>4</sub>.



**Figure 16** XPS survey spectrum of the designed RuO<sub>2</sub>/CoFe<sub>2</sub>O<sub>4</sub>.





**Figure 17** LSV curve of the designed RuO<sub>2</sub>/CoFe<sub>2</sub>O<sub>4</sub>.

**Table S1** Element content of Fe, Co Ru and P in 1M KOH after the hydrolysis determined using ICP.

State of solution	Element	the element content of the solution (mg/L)	Diluted multiples	Sample element content (mg/L)
<b>initial</b>	Fe	< 0.02	10	< 0.2
<b>final</b>	Fe	< 0.02	10	< 0.2
<b>initial</b>	Co	< 0.02	10	< 0.2
<b>final</b>	Co	0.03	10	0.31
<b>initial</b>	Ru	< 0.02	10	< 0.2
<b>final</b>	Ru	< 0.02	10	< 0.2
<b>initial</b>	P	< 0.02	10	< 0.2
<b>final</b>	P	7.59	10	75.94

**Table S2** Comparison of the electrocatalytic performances for HER in 1M KOH.

Catalysts	Electrolyte	Overpotential (mV) 10 mA cm <sup>-2</sup>	Tafel slope (mV dec <sup>-1</sup> )	Reference
CoFeO <sub>x</sub> H <sub>y</sub> -Ru/P-CoFe <sub>2</sub> O <sub>4</sub> /IF	1 M KOH	22@20 mA cm <sup>-2</sup>	43.2	This work
RuO <sub>2</sub> @C	1 M KOH	20	46	<i>Nano Energy</i> 2019, 55, 49-58
RuCoP	1 M KOH	23	37	<i>Energy Environ. Sci.</i> 2018, 11, 1819-1827
Ru <sub>2</sub> -GC	1 M KOH	25	65	<i>ACS Catal.</i> 2018, 8, 11094-11102
Ru@SC-CDs	1 M KOH	29	57	<i>Nano Energy</i> 2019, 65, 104023
Ru@CN	1 M KOH	32	53	<i>Energy Environ. Sci.</i> 2018, 11, 800-806
Ru/CP	1 M KOH	35	50	<i>Nanoscale</i> 2017, 9, 16616-16621
Ru-MoS <sub>2</sub> /CNT	1 M KOH	50	62	<i>Adv. Sci.</i> 2019, 6, 1900090
Ru-ZIF-900	1 M KOH	51.6	78.4	<i>J. Mater. Chem. A</i> 2020, 8, 3203-3210
RuP <sub>2</sub> @NPC	1 M KOH	52	69	<i>Angew. Chem., Int. Ed.</i> 2017, 56, 11559-11564
Ru <sub>0.33</sub> Se @ TNA	1 M KOH	57	50	<i>Small</i> 2018, 14, 1802132
Cu <sub>2-x</sub> S@Ru	1 M KOH	82	48	<i>Small</i> 2017, 13, 1700052

**Table S3** Comparison of the electrocatalytic performances for HER in alkaline medium.

Catalysts	Electrolyte	Overpotential (mV) at 100 mA cm <sup>-2</sup>	Reference
CoFeO <sub>x</sub> H <sub>y</sub> -Ru/P-CoFe <sub>2</sub> O <sub>4</sub> /IF	1 M KOH	54.4	This work
RuO <sub>x</sub> -Ni(OH) <sub>2</sub> /NF	1 M KOH	61.7	<i>Electrochimica Acta</i> 2020, 356, 136732
RuP(S-RP/C)	1 M KOH	71	<i>Adv. Mater.</i> 2018, 30, 1800047
Ru-HPC	1 M KOH	~80	<i>Nano Energy</i> 2019, 58, 1-10
Pt	1 M KOH	87	<i>Small Methods</i> , 2020, 4, 1900796

Fe <sub>0.5</sub> Co <sub>0.5</sub> P	1 M KOH	98	<i>Energy Environ. Sci.</i> , 2018,11, 2246-2252
Ru NP/C	1 M KOH	~100	<i>Adv. Energy Mater.</i> 2018, 8, 1801698
Pt <sub>SA</sub> -Co(OH) <sub>2</sub> @Ag NWs	1 M KOH	104	<i>Energy Environ. Sci.</i> , 2020,13, 3082-3092
Ni-MoO <sub>2</sub> -400 NWs-CC	1 M KOH	105	<i>J Mater Chem A</i> 2017, 5, 24453-24461
Co P/CNT	1 M KOH	109	<i>Nat. Commun.</i> 2016, 7, 10771
Pt <sub>SA</sub> -NT-NF	1 M KOH	110	<i>Angew. Chem. Int. Ed.</i> , 2017, 56, 13694-13698
Pt/Ni(HCO <sub>3</sub> ) <sub>2</sub>	1 M KOH	120	<i>Angew. Chem. Int. Ed.</i> 2019, 58, 5432
Pt-Co(OH) <sub>2</sub> /CC	1 M KOH	122	<i>ACS Catal.</i> , 2017, 7, 7131-7135
Pt SNs -MoO <sub>2</sub> NRs	1 M KOH	135	<i>Chem. Eng. J.</i> 2022, 427, 131309
V-CoP/CC	1 M KOH	138	<i>Chem. Sci.</i> 2018, 9, 1970
CoP/Ni <sub>5</sub> P <sub>4</sub> /CoP	1 M KOH	140	<i>Energy Environ. Sci.</i> 2018, 11, 2246
Ni-BDT	1 M KOH	150	<i>Chem</i> 2017, 3, 122-133
Ni <sub>3</sub> N/Pt	1 M KOH	170	<i>Adv. Energy Mater.</i> 2017, 7, 1601390
Ru-Fe <sub>3</sub> O <sub>4</sub> @FeNi-LDH-NF	1 M KOH	212	<i>Dalton Trans.</i> , 2021, 50, 13951-13960
RuCo@NC	0.1 M KOH	218	<i>Nat. Commun.</i> 2017, 8, 14969

**Table S4** Comparison of the electrocatalytic performances for HER in neutral.

Catalysts	Electrolyte	Overpotential (mV) at 10 mA cm <sup>-2</sup>	Reference
CoFeO <sub>x</sub> H <sub>y</sub> -Ru/P-CoFe <sub>2</sub> O <sub>4</sub> /IF	1 M PBS	30.1@ 50 mA cm <sup>-2</sup>	This work
Rh <sub>2</sub> P	1 M PBS	38	<i>Adv. Energy Mater.</i> 2018, 8, 1703489
Ru, Cr <sub>2</sub> O <sub>3</sub> /NG	1 M PBS	53	<i>RSC Adv.</i> , 2021,11, 6107-6113
RuP <sub>2</sub> @NPC	1 M PBS	57	<i>Angew. Chem. Int. Ed.</i> 2017, 56,11559
CC@WO <sub>3</sub> /Ru-450	1 M PBS	64	<i>Chem. Eng. J.</i> 2022, 430, 132953

Ru/OMSNNC	1 M PBS	70	<i>Adv. Mater.</i> 2021, 2006965
Ru <sub>SA</sub> -N-Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	1 M PBS	81	<i>J. Mater. Chem. A</i> 2020, 8, 24710–24717
Ru/MEOH/THF	0.1 M PBS	83	<i>Chem. Commun.</i> , 2017, 53, 11713
Rh <sub>2</sub> P/NPC	0.2 M PBS	119	<i>J. Mater. Chem. A</i> 2020, 8, 25768–25779

**Table S5.** Comparison of the electrocatalytic performances overall water splitting

Catalysts	Electrolyte	Cell Voltage (V) at 10 mA cm <sup>-2</sup>	Reference
CoFeO <sub>x</sub> H <sub>y</sub> -Ru/P-CoFe <sub>2</sub> O <sub>4</sub> /IF	1 M KOH	1.49	This work
Rh SAC-CuO NAs/CF	1 M KOH	1.51	<i>Nano Lett.</i> 2020, 20, 5482-5489
Ru-NiCoP/NF	1 M KOH	1.515	<i>Appl. Catal. B: Environ.</i> 2020, 279
Ru/NiFe LDH-F/NF	1 M KOH	1.53	<i>Nanoscale</i> 2020, 12, 9669-9679
Pt-CoS <sub>2</sub> /CC	1 M KOH	1.55	<i>Appl. Catal. B: Environ.</i> 2019, 249, 91-97
RuO <sub>2</sub> /N-C	1 M KOH	1.55	<i>J. Mater. Chem. A</i> 2018, 6, 1376-1381
IrTe <sub>2</sub>	1 M KOH	1.56	<i>Adv. Funct. Mater.</i> 2020, 30, 2004375
Ru <sub>1</sub> Ni <sub>1</sub> -NCNFs	1 M KOH	1.564	<i>Adv. Sci.</i> 2020, 7, 1901833
Ru <sub>1</sub> Co <sub>2</sub> NPs	1 M KOH	1.59	<i>ACS Appl. Energy Mater.</i> 2020, 3, 1869-1874

